

Epitaxial Lattice Matching and the Growth Techniques of Compound Semiconductors for their Potential Photovoltaic Applications

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ABSTRACT

This paper presents the recent advances in semiconductor alloys for photovoltaic applications. The two main growth techniques involved in these compounds are metal organic chemical vapor deposition (MOCVD) and molecular beam epitaxy (MBE), that has also been discussed. With these techniques, hetero-structures can be grown with a high efficiency. A combination of more than one semiconductor like GaAs, InGaAs and CuInGaAs increases the range of their electrical and optical properties. A large range of direct band gap, high optical absorption and emission coefficients make these materials optimally suitable for converting the light to electrical energy. Their electronic structures reveal that they are highly suitable for photovoltaic applications also because they exhibit spin orbit resonance and metal/semiconductor transitions. The dissociation energy has also been discussed in reference to the increased stability of these compounds.

Keywords: Compound semiconductors, hetero-junctions, lattice mismatching, photovoltaic application, variable band gap

1 Introduction

The various semiconductor materials from Group II to group VI are of interest to solar photovoltaic since they possess direct band gap. Band gap energy is an important parameter for the choice of the semiconductor material. The lattice parameters and the band gaps can be tailored for the ternary and quaternary compound semiconductors. It has been observed that the ternary XYZ_2 chalcopyrites ($X = \text{Cu}$, $Y = \text{In}/\text{Ga}$, Al , $Z = \text{S}/\text{Se}/\text{Te}$) have diversity in their structural, electrical and optical properties making them materials of interest for the generation of electrical energy through the solar cells [1]. Their efficiencies are as high as 19.2% with a Cu (In, Ga) Se_2 (CIGS) absorber with a band gap value of 1.0- 1.7 eV. The addition of Ga tends to increase the band gap up to 1.15 eV. The lattice mismatching for GaInP/GaInAs/Ge, which is a three junction solar cell, has also been achieved with efficiency of 37.3% at 175 suns, 25°C under the standard AM 1.5D and terrestrial spectrum [2], [3].

Due to complex defect structures of II-VI semiconductors and thus the fabrication of the extrinsic type materials has been gradual. *Liquid Phase epitaxy*, *Chemical Vapor Deposition*, (CVD), *Metal Organic Chemical Vapor Deposition* (MOCVD), *Molecular Beam Epitaxy* (MBE) are the most applicable techniques which have been discussed in this review.

These compound semiconductors find applications for light sources and the detectors. They find potential applications in MESFETs, LEDs, HBTs, FETs, Optoelectronic devices, Infrared Imagers, thermo-photovoltaics (TPV), Ohmic Contacts, Radiation detectors, photo-refractive gratings, solar cells [1], [4]. The HgCdTe and ZnSse of the II-VI groups are of technological importance in the lasers, LEDs, photo detectors for the far infrared detectors and blue visible emitters.

2 Compound Semiconductors

Compound semiconductors are the semiconductors that are made from two or more elements. These are the next generation of

semiconductors. Most of these compound semiconductors are from Group III and V of the periodic table (GaAs, GaP, InP etc). The group II and VI based compound semiconductors are CdTe, ZnSe. Compound semiconductors can also be made from within the same group IV such as SiC. The properties like wide band gap, high mobility and direct band gap make them potential materials for the application in different areas. These semiconductors are difficult to grow as compared to Si but their special properties make them better candidates for technological applications. They are used in power amplifiers for smartphones and other wireless devices, light sources for DVDs and Blu-rays, LEDs, solar batteries, and solar cells and gyro stabilizers in satellites.

Some of their characteristic properties are high electron mobility for high speed operations, ability to generate high frequency microwave signals, low voltage operation (for lighter and longer lasting, low voltage batteries used for portable batteries), light emission-both visible and infrared for lasers and LEDs, sensitivity to light and magnetism for their applications in photo-detectors and sensor applications. They offer resistance to heat and radiation and thus useful for space applications such as solar cells. The compound semiconductors also find applications in the field of communication,

particularly in Optoelectronics and HF systems (800MHz -80 GHz).

In the photo-voltaics, energy conversion, excitation of electrons to higher potential energy level and the separation of charges from the excited state occurs for the generation of electricity. For this the semiconductors in their elemental and compound states are suitable, since their conductivity can be controlled by the presence of impurities and optical excitation. The Si based solar cells have an indirect band gap in the crystalline form. But their efficiencies have been reported to be below the III-V compounds. The amorphous Si cells are promising and may be exploited for their potential applications in the area of photovoltaics. It has been observed that most of the III-V compound semiconductors are zinc-blende and wurtzite structures and the variation in composition for band gap modification is useful in designing high efficiency solar cells especially for space applications.

These materials have high dissociation energies increasing their stabilities. They become radiation resistant and are appropriate for space exploration. Both direct and indirect band gap semiconductors are used for solar cell applications. However, the indirect band gap materials require a photon and a phonon for the excitation of an electron from the valence to conduction band.

Table 1: Essential parameters of some elemental and compound semiconductors

Element/ Compound	Lattice Constant (Å)	Band Gap (eV)	Band Symmetry	Density of States (cm ⁻³)	
	at 300K	at 300K		Nc	Nv
Ge	5.646	0.66	Direct	1.05 x 10 ¹⁹	3.92 x 10 ¹⁸
Si	5.430	1.12	Indirect	2.8 x 10 ¹⁹	1.02 x 10 ¹⁹
GaP	5.4512	2.24	Indirect	3.2 x 10 ¹⁹	1.8 x 10 ¹⁹
GaAs	5.653	1.42	Direct	4.7 x 10 ¹⁷	9.0 x 10 ¹⁸
GaSb	6.0959	0.72	Direct	2.1 x 10 ¹⁷	1.8 x 10 ¹⁹
ZnS	5.420	3.68	Direct	---	---
ZnSe	5.668	2.71	Direct	---	---
ZnTe	6.103	2.26	Direct	---	---
CdS	5.832	2.42	Direct	---	---
CdSe	6.050	1.70	Direct	---	---
CdTe	6.482	1.56	Direct	---	---
GaN	3.36	3.32	Direct	1.2 x 10 ¹⁸	4.1 x 10 ¹⁹
AlN	6.2	5.74	Indirect	6.3 x 10 ¹⁸	4.8 x 10 ²⁰
Al _x Ga _{1-x} As [5]	5.65381	1.5 to 2.0	Direct	---	---

This decreases the probability of light absorption and consequently a thicker layer of this type of material will be required for the absorption of complete solar spectrum [6]. As it can be seen from Table 1, the semiconductors are used in their elemental, binary, ternary and quaternary configurations. The ternary alloys are alloys of two binary compounds and one common element. They provide a continuous range of band gaps, and their properties such as effective mass also vary quadratically with a variation in alloy fraction. The substrates of these compounds are always binary and are found on discrete value of lattice constant. So, growing hetero-structures requires different E_g layers having the same lattice constant "a".

The III-V group semiconductors that lattice matched to GaAs (i.e AlGaAs, InGaAlP) and InP (i.e InGaAsP) have been extensively studied for their technological applications. GaAs and InP are have a compatibility of getting alloyed with Al, Sb and give rise to ternary and quaternary compounds. $Ga_xAl_{1-x}As$ is of technological importance since GaAs and AlAs form a solid solution in the range ($0 \leq X \leq 1$) with a small variation in the lattice parameter. This is very useful for the fabrication of quality multi layers. Within the range of ($0 \leq x \leq 0.35$) a direct band gap is obtained whereas variation in fraction with $x \geq 0.35$ results in indirect band gap. The strain between the layers or the lattice mismatch is a constraint as the compositional change in the alloy also varies the lattice constant [7], [8].

The InP/InGaAs hetero-structure is sensitive to 1.55 μm of wavelength. A bad gap of 0.75 eV is a result of the lattice matching constraint to the InP substrate. The lattice mismatch manifests itself in the form of low quantum efficiency in optoelectronic devices [9], [10].

The incorporation of N atoms in the III-V group compound semiconductors like GaN_xSb_{1-x} , indicates a large band gap reduction. This has been correlated with the fact that nitrogen introduces localized levels in the band structures that interact with the conduction band and result into the formation of non-parabolic sub bands [11].

3 Growth Techniques

The II-VI semiconductors have complex defect structures and thus the fabrication of the extrinsic type materials has been gradual [12]. Problems also arise due to certain properties of these materials. The ionic nature of the bonds between the anions and the cations is challenging in the growth of the crystals. The doping of mercury in II-VI group-based semiconductors gives rise to toxicity and high pressures are created. It is also difficult to obtain the crystals that are homogeneous in composition. The poor conductivity of these materials at the growth temperatures leads to crystal fluid concave shaped interface. The various growth techniques used are Epitaxial growth technologies and bulk crystal growth [13], [14]. The epitaxial growth techniques are useful in device applications. Stacking faults are observed in their structures. The formation of large number of the vacancies and defects affects the electrical properties of II-VI compound semiconductors. The most commonly used n-type dopants are sulphur, selenium, tellurium, tin, silicon, carbon, germanium and p-type dopants are zinc, beryllium, magnesium, cadmium, silicon, carbon, germanium. Si, C and Ge act as both n and p-type dopants that are capable of replacing the Ga or As atom in the crystalline structure [15]. Figure 1 shows relationship between Lattice Constants, Band gap energies and Band-gap wavelength of II-VI compounds [16].

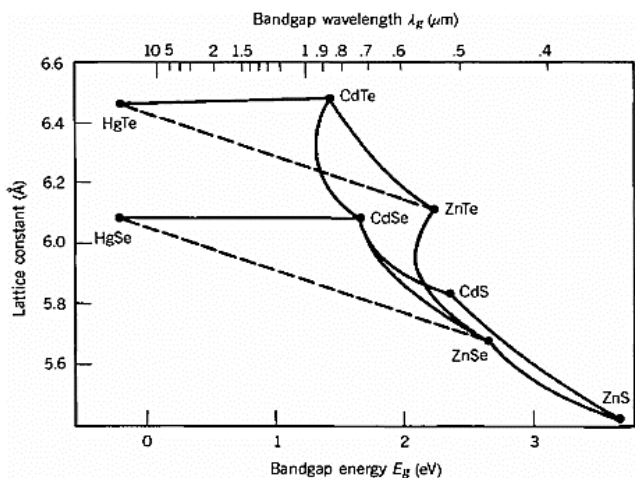


Figure 1: Lattice Constants, Band gap energies and Band-gap wavelength of II-VI compounds [16]

3.1 Epitaxial Growth Techniques

The term epitaxy originates from the Greek words, "epi" and "taxis" which mean to arrange upon. Epitaxy is an arrangement of atoms in a crystal formed on a substrate in a manner that the deposited layer is an extension of the substrate crystal structure. These techniques are of importance in electronic circuit fabrication as the devices that need to be fabricated need only few microns of dimension. It reduces the growth time, wafering cost and eliminates the wastages caused during growth, cutting, polishing etc. The major advantage of epitaxy is the uniformity in composition, controlled growth parameters and a better understanding of the growth. Epitaxial growth techniques are used for III-V and II to VI group compound semiconductors. The crystallinity matches that of the substrate even though the composition of the materials may differ (e.g. GaAlAs/GaAs, SiGe/Si, CdTe/GaAs). It involves the growth of one crystal on another with well related and matched orientations. Epitaxy are of two types namely *Homo-epitaxy* in which the thin crystal lattice layer is the same as that of the substrate. For example, Si film on Si substrate and the other type is *Hetero-epitaxy* in which the thin crystal lattice layer is different from that of the substrate as in GaAs on Si substrate. The Group III-V semiconductors exhibit good electronic and opto-electronic properties. In order to grow, single crystal thin films, closely matched epitaxial substrates are required mostly amorphous in nature. InP is useful and patterned InP are configured in the form of transistors, and photo-detectors on the amorphous SiO₂ substrates. The ternary and quaternary compounds like $Al_xGa_{1-x}As$ and $In_xGa_{1-x}As_yP_{1-y}$ are being prepared by this techniques since Ge, GaAs and AlAs demonstrate a matching in their lattice constants with difference in the band gaps as can be seen from Figure 2. Epitaxy offers a low growth rate of the semiconductors due to low temperatures used in this method. With this method, high purity compounds are obtained. It is also reported that they offer high values of mobility, life time and free mean path for the minority carriers [17]–[21]. The methods used for epitaxial

growth are *Liquid Phase epitaxy*, *Chemical Vapor Deposition*, (CVD), *Metal Organic Chemical Vapor Deposition (MOCVD)*, *Molecular Beam Epitaxy (MBE)* and are described under.

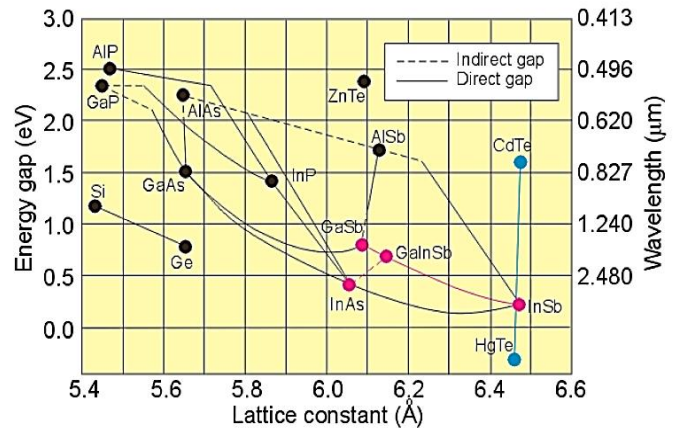


Figure 2: Relation between the lattice constant and the band gap of some compound semiconductors [22].

3.1.1 Liquid Phase Epitaxy (LPE)

Liquid Phase Epitaxy is a method of growing semiconductor crystal layers from the melt on solid substrates and it occurs at temperatures below the melting point of the deposited semiconductor. The process is not complex and easy to operate and offers large temperature range of growth. Figure 3 shows line diagram of Liquid Phase Epitaxy.

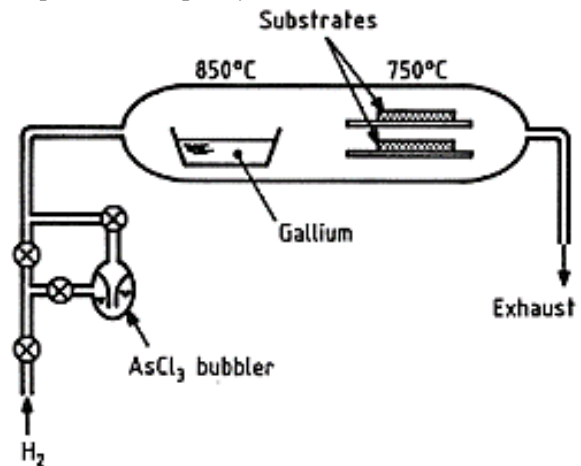


Figure 3: Line diagram of Liquid Phase Epitaxy [23]

A higher growth rate is possible with LPE which is about 0.1-1 μm/min. Since the growth takes place near the equilibrium, lesser defects are observed. So, the crystal quality is excellent. It is an inexpensive and fast technique. This technique also takes the advantage of the fact that impurity addition to the molten material lowers its melting

point. As a result, the molten compound can be kept at lower temperature in way that does not allow the seed to melt. Slow cooling of the melt results in the growth of the epitaxial layers. Epitaxial growth is controlled by the rate at which the temperature is lowered. For GaAs, the growth temperatures are in the range of 350°C – 900°C and relatively lower concentration of point defects have been reported. Indium-phosphide (InP) is also used as a substrate. It finds applications in LEDs and laser diodes.

Recent developments in the fabrication of the device structures like double hetero-structures (DH), laser diodes, pin photodiodes, avalanche photodiodes, Gunn diodes, integrated bipolar transistor-laser circuits, pin-FET photo-receivers, multi quantum well lasers have made liquid phase epitaxy a unique distinction amongst the other growth techniques.

3.1.2 Chemical Vapor Deposition (CVD)

This technique is useful for the growth of polycrystalline, amorphous and single crystalline thin films that are used in the fabrication of semiconductor-based devices and integrated circuits in which thin layers of semiconductors, metals and insulators are to be laid. Thin solid films of high epi-quality and high growth rates can be obtained from the vapor phase through a chemical reaction. Silicon dioxide, Silicon nitride and single crystal Silicon layers of controlled thickness and electrical properties can be grown by this method. The materials in their gaseous form are passed through a reaction chamber. They should be able to react near or on the surface of the substrate for the deposition to take place. The *halide CVD* process involves the introduction of group V as chloride (AsCl_3) over the metal Gallium highest purity GaAs is formed at temperatures of about 650-850°C (Figure 4). This heat energy dissociates the source materials in free radicals. UV radiation and Plasma energy is also used to heat the substrate. The growth rates by this method is about 1 $\mu\text{m/h}$. [24]–[26] and CVD can be carried out at atmospheric or low pressures. The important chemical reactions involved in this technique are:

i) **Pyrolysis**- Dissociation of the compound on applying the heat energy

- ii) **Photolysis**- Dissociation of the compound on applying the external energy
- iii) **Reduction**- A component of the compound gets free during the reaction to form a new compound with the lowering of the valency.

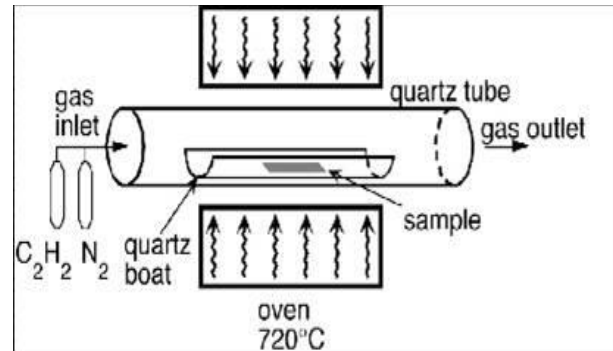


Figure 4: Chemical Vapor Deposition technique [27]

The *hydride CVD* involves the introduction of group V element as hydride (AsH_3) and it results in the formation of metal chloride, by passing HCl over the metal. This method is more toxic but offers flexibility in the growth of binary, ternary and quaternary III-V materials. A recent advancement is the introduction of a “double barrel reactor” in which a use of two parallel CVD systems that feed into a single growth Chamber. This allows the switching of the substrate from one CVD source to the other to form abrupt interfaces. It also removes the problem in growing abrupt hetero-structures where changing from one gas source to another would require pump-out and pre heat cycles which tend to generate interfacial defects [28].

3.1.3 Metal Organic Chemical Vapor Deposition (MOCVD)

It is a process that is used to produce high purity crystalline compound semiconducting thin films of the order of 1nm thickness on the substrates. The growth process is thermodynamically favorable. The dopants can be added in precisely controlled amounts in this process. It is used for the synthesis of III-V compound semiconductors which have high volatility, especially GaN based which can be processed into electronic, opto-electronics devices, high speed transistors etc. MOCVD has emerged as a leading candidate for commercial GaN device application in achieving super blue light emitting diodes. MOCVD is a

non-equilibrium technique which relies on vapor transport of the precursors and subsequent reactions of group III alkyls and group V hydrides in heated zone (Refer to Figure 5).

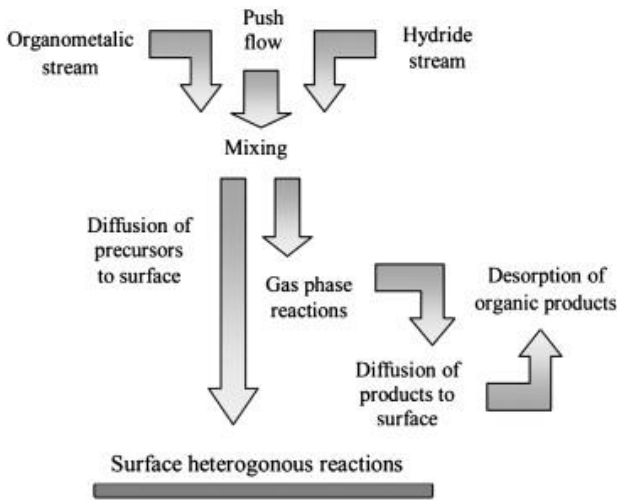
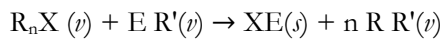


Figure 5: Flow diagram of the Metal Organic Chemical Vapor Deposition

This method involves the vaporization and transportation of the chemicals involved into a reactor where the chemical reaction takes place at temperatures around the room temperature and is thermally decomposed by hot susceptor and substrate to form the required film inside the reaction chamber.

The general equation that represents the MOCVD process is:



where R and R' are the methyl or ethyl radicals or hydrogen, X is the Group II or III metal, E is the Group V or VI element, $n=2$ or 3 and ν and s indicate the phase. The product RR' is carried away with the flush of hydrogen to the exhaust. For the growth of crystalline GaAs atoms onto the substrate, Arsine gas (AsH_3) is passed over Trimethyl gallium gas $Ga(CH_3)_3$, the heated semiconductor substrate. The heat breaks up the molecules and deposits the desired atoms on the surface layer by layer. The atoms bond to the substrate layer and a new crystalline layer is grown.

MOCVD has been found to give good results for phosphorous compounds as in InP where $TmIn$ is used as precursor. It is also widely used to produce the quantum well structures and multi-junctions. A new development is to grow a

selective area with laser beam dissociation. The dissociation of organo-metallic molecules results in selective area deposition. Photodetectors are fabricated with this technique [29]. However, the disadvantage of MOCVD is that it is difficult to monitor the growth rate exactly since no RHEED is possible due to higher pressure. Toxic gases are produced that need to be handled.

3.1.4 Molecular Beam Epitaxy (MBE)

Molecular beam epitaxy was developed by Alfred Cho and John Arthur at Bell Telephone Labs in the early 1970s, in an attempt to develop a new variety of high frequency and high temperature devices such as injection lasers, electro-optic phase modulators, photo cathodes and oscillators, as well as IMPATT diodes and optical waveguides [30]. Molecular beam epitaxy is a valuable tool in the development of electronic and optoelectronic devices since it provides sharp and clear interface between different materials of choices. The pioneering work was focused mainly on the preparation of $A_{III}B_V$ compound films like GaAs.

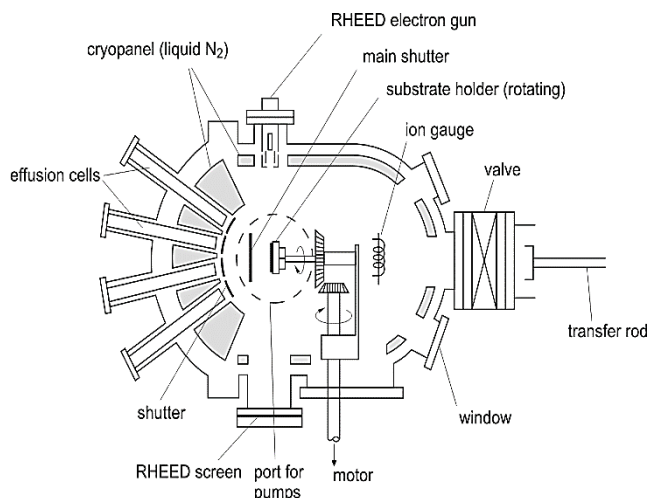


Figure 6: Schematic diagram of Molecular Beam epitaxy [31]

Molecular beam epitaxy technique is used to grow high purity epitaxial layers of compound semiconductors of III-V group. Figure 6 shows an outline of the molecular beam epitaxy set up. In this technique the constituent elements of the semiconductor are deposited on heated crystalline substrate and thin epitaxial layers are formed. The atoms of the beam are then adsorbed by the sample surface. The interaction

of the atoms to be adsorbed and the atoms of the sample surface depends upon the type of atoms, substrate and its temperature. MBE is used for producing super-lattice structures that consist of thin layers with thickness as low as 10 Å. It provides a control on compositional profiles since the impurities are evaporated onto the growing film through separate sources. Hence the doping profile can also be controlled and varied with a good spatial resolution. Metal organic group III precursors, gaseous group V hydride of high purity in ultra-high vacuum environment, are usually used as the molecular beam. An Ultra high vacuum (UHV) is essential for MBE with a total pressure within the deposition chamber lower than 10^{-10} Torr. The rate of gas evolution from the materials in the chamber have negligible partial pressures as compared to the sources. The source evaporates at approximately 600°C (Solid source MBE) or gets cracked into elemental form (gas-source MBE). Pyrolytic boron nitride based crucibles are used for they provide a chemical stability upto 1400°C, molybdenum and tantalum are used for shutters. The operation time is approximately 0.1 s and it is shorter than the time needed to grow one monolayer typically of 1-5 s [30]. The chamber is heated for 200°C for 24 hours before attaining the UHV. The evaporated elements do not interact with each other or with the vacuum chamber gases until they impinge the substrate due to their long mean free paths, that are involved in the deposition process. Nitrogen gas having a molecular weight of 28.01 has a mean free path of 6.044×10^{-8} m. So, the element travels along a straight line and gets deposited on the substrate which is heated at about 400°C. The UHV conditions in the environment and control of the quality of the source materials allows films of higher purity as compared to the other growth techniques. This technique provides a clean growth environment, a provides a precise control of the beam fluxes and growth condition by changing the nature of the incoming beam just by opening and closing the mechanical shutters. Reflection high energy electron diffraction (RHEED), a characterization tool requires UHV environment which is available in MBE. RHEED can be used to monitor the surface

reconstruction due to its shallow incidence angle. It is highly compatible with other high vacuum thin film processing methods like metal evaporation, ion beam milling and ion implantation.

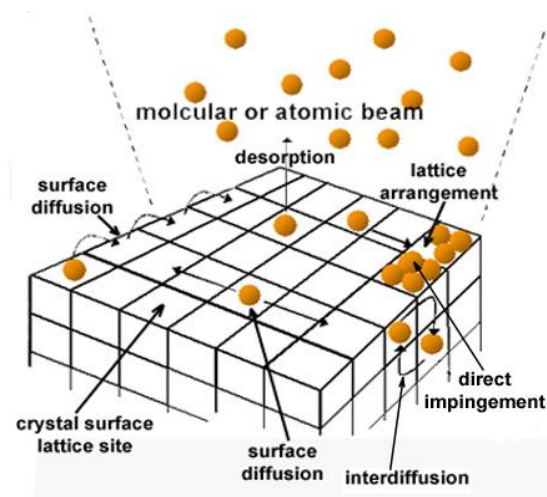


Figure 7: Schematic illustration of the surface processes occurring during MBE growth [30]

The processes involved during the epitaxial growth can be summarized as: a) adsorption of the evaporated atoms or molecules impinging on the surface; b) surface diffusion and dissociation of the adsorbed atoms; c) atoms arrangement into the crystal lattice; d) thermal desorption of the species not arranged into the lattice [32]. The adsorbed molecules (or atoms) first combine to reach a critical size, and then become stable nuclei (or clusters) on the substrate. The main factors affecting the critical size are the substrate temperature, the arrival rates and the combining capacity with the substrate. Subsequent molecules (or atoms) may condense directly onto these nuclei, decreasing the rate of formation of new nuclei and increasing the size of existing ones to form islands that may coalesce upon surface contact (Refer to Figure 7). One of the recent efforts for device fabrication has been to prepare compound semiconducting materials with band energies from 0.75 eV to 1.55 eV that correspond to wavelengths from 1.65 μm to 0.8 μm . The long wavelength end covers the lowest transmission loss of the present fiber optic communication systems [33].

The advantage of MBE is that it permits growth of crystalline layers at temperatures where solid state diffusion is negligible. The deposition

species require little energy to migrate along the substrate surface to crystalline bonding site. The incorporation of impurity dopant is possible by having an additional source of dopant. Consequently, it is a versatile technique for growing elemental and compound semiconductor films. It can be used to generate complicated doping profiles as it regulates the amount of the dopants. The advantages of this growth technique are that since it is low temperature technique, hence it finds application in VLSI. No chemical reactions are involved as it is based on the evaporation of Si and the dopants. However, the Molecular Beam Epitaxy suffers of lower yield compared to other techniques like Liquid Phase Epitaxy, Metal Organic Vapor phase deposition (MOCVD), Sputtering and Plasma Enhanced Chemical Vapor Deposition (PECVD) due to a lower speed process and dimension wafer capability. The growth rate is also very slow i.e 0.01-0.3 gm/min and the process are very expensive.

4 Conclusion

Now a days, III to V group semiconductors are being tailored to fine scales by advanced micro and nano technology. These material form homogeneous solid solutions or alloys since they are soluble with each other. The unconventional alloys formed from II-VI group semiconductors like GaInNAs are of importance and are being investigated with an intent of increasing the solar cell efficiency. Hence, the future photovoltaic technology can be improved in terms of efficiency by researching on the band tailored combinations of II-VI group semiconductors. The future technology shall be revolutionized by the compound semiconductors. In electronics industry, they will enable 5G to robotics, more efficient renewable energy and autonomous vehicles are expected to be developed. They will also underpin Internet of things (IoT) which is 5G dependent and moreover these semiconductors are power efficient which is an essential feature for the battery operated devices. They have unique optical properties and so they find applications in sensors that will deliver new imaging techniques for connected cars, healthcare etc.

The epitaxial growth techniques are of significance to the compound semiconductors especially for their potential applications in the field of semiconductor microtechnology. The development of advanced technology is based on the improvement of the base quality, which is possible by the growth techniques discussed above. They are used to deposit thin films of a few micrometers onto a flat wafer. MBE is used extensively in the growth of these semiconductors for their applications in semiconductor lasers and photodetectors. It is in particular used to grow semiconductors of group from II to VI, single crystal metal films, and insulators. The epitaxial growth techniques provide uniformity of growth rate and alloy composition. Thin multilayered structures can be also grown which have promising applications in the field of technology. The doping profiles can be controlled on a nanometer scale.

A problem utilizing these materials for their potential application is the difficulty in controlling their conductive type due to the native defects that occur which may have donor, acceptor or amphoteric character. They act as compensating centers. These defects also have a tendency of combining with dopant impurities to form complexes. Therefore, a significant improvement in the understanding of the fundamental physical and chemical properties needs to be achieved.

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